

Optical Anisotropy of Self-assembled Nanostructure in Glass

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Abstract: Femtosecond laser direct writing of form birefringence originated from self-organized nanostructure in glass is reviewed. Its application to rewritable five-dimensional optical data storage is also demonstrated.

OCIS codes: (320.7120) Ultrafast phenomena; (220.4241) Nanostructure fabrication

1. Introduction

Material processing with ultrafast lasers has recently attracted considerable interest [1] due to a wide range of applications including laser surgery [2], 3D micro- and nano-structuring [3]. An intriguing phenomenon which currently attracts a lot of interest is self-assembly of periodic nanostructures in the direction perpendicular to the light polarization [3-5]. Uniaxial birefringence observed after femtosecond laser irradiation of silica glass [6] has been explained by induced nanogratings and referred as self-assembled form birefringence [7, 8]. Self-organization process has been interpreted in terms of the interference of electron plasma waves resulting in electron concentration modulation, followed by freezing of the interference pattern by structural change in glass. Self-assembled nanostructure, which is composed of the periodic modulation of oxygen defects, has initially evolved from residual birefringence originated from internal stress distribution due to thermal distribution with steep gradients [9]. Evolution of optical anisotropy in glass can be controlled as a function of interpulse time due to thermal accumulation. Especially for the longer interpulse interval corresponding to the lower thermal accumulation effect, in spite of the same pulse energy, the evolution of laser-induced-damage is obviously different depending on the polarization direction due to local heating with strong polarization-dependence [10]. Such phenomenon can also be observed in the plasma emission during laser irradiation indicating the polarization-dependent energy transfer in laser produced plasma. The stress accumulation and the oxygen defect generation depend on the polarization direction, which could be explained by the anisotropy of electron plasma absorption for p and s polarizations at the oblique, moving with the speed of light interface produced by the pulse with tilted intensity front. Such anisotropy affects the reciprocal evolution of the material modification in isotropic medium, finally exhibits a non-reciprocal quill writing effect [11] revealing strong dependence of the material modification on the orientation of writing direction relative to the direction of pulse front tilt [12]. Apart from the basic understanding, applying this technique to optical storage, we can achieve the rewritable five dimensions of optical memory (i.e. three dimensions of space (XYZ), polarization direction (E), and phase retardance (δ)).

2. Experimental

The experiments were performed using a mode-locked, regeneratively amplified Ti: Sapphire laser system (Coherent; RegA 9000), operating at 800 nm with 70 fs pulse duration and 250 kHz repetition rate. Repetition rate of the laser was set to 250 kHz corresponding to the interpulse interval τ_{int} of 4 μs . The femtosecond laser beam was focused via a microscope objective (Nikon; LU Plan Fluor, 100 $\times$ 0.90 N.A.) at a depth of about 100 μm below the surface of silica glass sample. Typical pulse energy was about 1.0 μJ and the beam power measured after microscope objective was independent on the orientation of light polarization. Previously we have observed the periodic nanostructure was induced in the orthogonal direction to the laser light polarization [3].

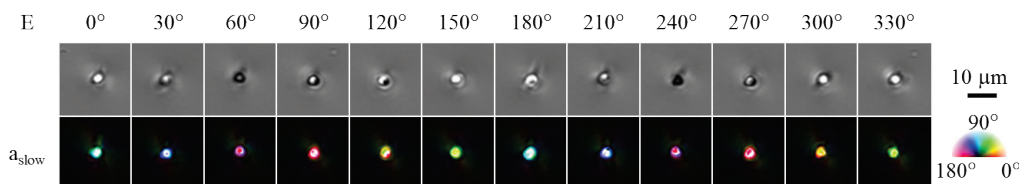


Fig. 1. Characteristic micrographs of localized birefringence, induced by focused femtosecond laser with various polarization direction (E), taken with optical (upper row) and polarization (lower row) microscope (pseudocolor indicates direction of the slow axis, a_{slow} , see polar legend).

Such nanograting structures, consisting of periodic oxygen defect regions, are responsible for form birefringence [7]. The direction of slow axis of birefringence (a_{slow}), which is always perpendicular to the writing light polarization (E), can be controlled by rotating polarization (Fig. 1). Erasure of nanostructure requires sufficiently high temperatures as the induced oxygen defects are maintained up to the glass transition temperature T_g of ~ 1200 °C [9]. The interpulse time τ_{int} and the number of pulses N_{pulse} were controlled by field programmable gate array (National Inst.; FPGA module 7811R) produced the trigger pulses. In order to reveal the growth mechanism of the anisotropic nanostructure, we have investigated the evolution of birefringence varying repetition rate (or the interpulse time τ_{int}) and exposition (or the number of pulse N_{pulse}) at the same pulse energy. After writing, the structures were inspected under an optical microscope and a polarized optical microscope. In the characterization, a confocal Raman spectrometer (Tokyo Instruments; Nanofinder 30) was used for structural identification of the irradiated regions. The localized birefringence was measured using a polarized optical microscope (CRi; LC-Polscope). By using the spectropolarimeter (Tokyo Instruments; Poxi-spectra), we have also observed wavelength-dependence of the birefringence of the modified region.

3. Results and Discussion

We experimentally demonstrate an application of photo-induced nanograting in the rewritable five-dimensional optical storage. To demonstrate the potential of this technology we saved the “Japanese woodprint of Mount Fuji” within a 2.5×1.6 mm square inside fused silica (Fig. 2). Printing of this image takes only about 10 minutes. Reading of the recorded information was realized with the cross-Nicole configuration (Fig. 2(b)). Basic working principle of this microscope is the following, a nearly monochromatic (wavelength: ~ 546 nm, bandwidth: ~ 10 nm) circularly polarized light illuminates the sample, then it is passed through a liquid crystal compensator, which modulates light polarization, and fixed analyzer before reaching a detector. As a result this multi-colored image corresponding to different slow axis orientation of the form birefringence was clearly observed (Fig. 2(c)). Simultaneously the phase retardance can be also read out (Fig. 2(d)). Due to the slight configuration errors in writing or the detection systems and limited sensitivity of CCD detector used for reading, the resolutions of the azimuth angle and the phase retardance were respectively less than 4.7° and 5 nm. By applying these techniques we have demonstrated the rewritable optical data storage in “five-dimensions” (3 coordinates, slow axis direction and retardance) with a 300 Gbit/cm³ capacity corresponding to the 10 times of the usual 12 cm BlueRay disk capacity. We anticipate that these results will motivate the development of rewritable 5D optical storage devices.

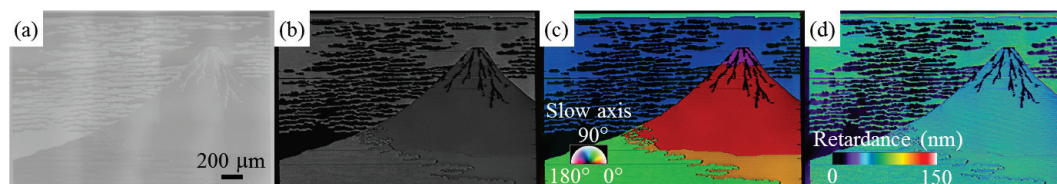


Fig. 2. Images of “Japanese woodprint of Mount Fuji” taken with optical (a) and polarization (b) microscopes. Actual size of the structure is 2.5×1.6 mm. Pseudo-color images of azimuth angle (c) and retardance (d) are also shown.

4. References

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